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PECASE- RESONATLY-ENHANCHED LANTHANIDE EMITTERS FOR SUBWAVELENGTH-SCALE, ACTIVE Photonics

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14. ABSTRACT

With the support of this grant, we: (1) developed energy-momentum spectroscopy, a new angle-resolved spectroscopic technique that allows for the determination of the strength, order, and orientation of electronic transitions in quantum emitters, (2) leveraged the spectrally distinct electric and magnetic dipole transitions in Eu3+:Y2O3 to tune emission spectra and to modulate emission at sub-lifetime speeds, (3) performed detailed quantum-mechanical calculations in the intermediate coupling regime to identify strong magnetic dipole emission lines for future experimental studies, (4) informed by these calculations we used energy-momentum spectroscopy to characterize electric and magnetic dipole contributions to Dy3+:Y2O3 and Tm3+:Y2O3 emission, (5) demonstrated spectral tuning of the highly mixed ED/MD telecom emission from Er3+:Y2O3 by varying the distance to a gold mirror with a Y2O3 spacer layer, (6) realized all optical modulation of Er3+:Y2O3 (at over 3 orders of magnitude faster than its excited state lifetime) by leveraging the ultra-fast VO2 insulator-metal phase transition, and (7) expanded energy-momentum spectroscopy to a larger range of quantum emitters by removing the entrance slit of the imaging spectrograph and using convex optimization to extract information from the multiplexed image, thus greatly increasing optical throughput.

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Archival publications (published) during reporting period:

[1] Christopher M. Dodson and Rashid Zia, "Magnetic dipole and electric quadrupole transitions in the trivalent lanthanide series: Calculated emission rates and oscillator strengths." *Physical Review B* **86** (2012), 125102, DOI: 10.1103/PhysRevB.86.125102

[2] Tim H. Taminiau, Sinan Karaveli, Niek F. van Hulst, and Rashid Zia, "Quantifying the magnetic nature of light emission". *Nature Communications* **3** (2012), 979, DOI:10.1038/ncomms1984

[3] Christopher M. Dodson, Jonathan A. Kurvits, Dongfang Li, and Rashid Zia. "Wide-angle energy-momentum spectroscopy". *Optics Letters* **39** (2014), 3927, DOI:10.1364/OL.39.003927

[4] Jon A. Schuller, Sinan Karaveli, Theanne Schiros, Keliang He, Shyuan Yang, Ioannis Kymissis, Jie Shan, and Rashid Zia. "Orientation of luminescent excitons in layered nanomaterials". *Nature Nanotechnology* **8** (2013), 271–276, DOI:10.1038/nnano.2013.20.

[5] Christopher M. Dodson, Jonathan A. Kurvits, Dongfang Li, Mingming Jiang and Rashid Zia. "Magnetic Dipole emission of Dy^{3+}:Y_2O_3 and Tm^{3+}:Y_2O_3 at near-infrared wavelengths". *Optical Material Express* **4** (2014), 2441-2450, DOI:10.1364/OME.4.002441

[6] Dongfang Li, Mingming Jian, Sebastien Cueff, Christopher M. Dodson, Sinan Karaveli, and Rashid Zia, "Quantifying and controlling the magnetic dipole contribution to 1.5μm light emission in erbium-doped yttrium oxide". Physical Review B **89** (2014), 161409, DOI:10.1103/PhysRevB.89.161409

[7] Sinan Karaveli and Rashid Zia, "Strong enhancement of magnetic dipole emission in a multilevel electronic system". *Optics Letters* **35** (2010), 3318-3320, DOI:10.1364/OL.35.003318

[8] Sinan Karaveli and Rashid Zia, "Spectral Tuning by Selective Enhancement of Electric and Magnetic Dipole Emission". *Physical Review Letters* **106** (2011), 193004, DOI:10.1103/PhysRevLett.106.193004

[9] Sinan Karaveli, Aaron J. Weinstein, and Rashid Zia. "Direct modulation of lanthanide emission at sub-lifetime scales". *Nano Letters* **13** (2013), 2264–2269, DOI:10.1021/nl400883r.

[10] Sebastien Cueff, Dongfang Li, You Zhou, Franklin J. Wong, Jonathan A. Kurvits, Shriram Ramanathan, and Rashid Zia. "Sub-lifetime modulation of erbium light emission at telecom wavelengths using VO_2 phase change", submitted.

[11] Sebastien Cueff, Joan Manel Ramirez, Jonathan A. Kurvits, Yonder Berencen, Rashid Zia, Blas Garrido, Richard Rizk, and Christophe Labbe. "Electroluminescence efficiencies of erbium in silicon-based hosts". *Applied Physics Letters* **103** (2013), 191109, DOI:10.1063/1.4829142.

[12] Dongfang Li, Nabil M. Lawandy, and Rashid Zia, "Surface phonon-polariton enhanced optical forces in silicon carbide nanostructures". *Optics Express* **21** (2013), 20900–20910, DOI:10.1364/OE.21.020900.

OBJECTIVE: The objective of this PECASE program was to develop active nanophotonic devices that leverage the electric *and* magnetic dipole transitions in lanthanide ions to realize a new class of subwavelength-scale optical sources, switches, and detectors.

BACKGROUND: Although it is often assumed that all light-matter interactions at optical frequencies are mediated by electric dipole transitions, strong optical frequency magnetic dipoles do exist. In fact, we see magnetic dipole emission every day from the many lanthanide ions (such as trivalent erbium and europium) that help to illuminate everything from fluorescent lighting to telecom fiber amplifiers. Our detailed quantum mechanical calculations have revealed over 1,900 magnetic dipole emission lines in the ultraviolet to near-infrared range between 300 nm and 1700 nm.[1] Nevertheless, most applications have overlooked the device implications of magnetic dipole transitions throughout the visible and near-infrared regime.

APPROACH: Despite similar radiation patterns, magnetic and electric dipole emitters have different symmetries with respect to polarization and phase. Thus, in an inhomogeneous environment, we can tailor interference effects and the local density of optical states to selectively enhance either electric or magnetic dipole emission.

Our work illustrates how the naturally occurring magnetic dipole transitions of lanthanide ions provide a new degree of design freedom for active photonic devices. Specifically, we have shown how the different symmetries of electric and magnetic dipoles can be exploited to quantify[2-6] and control[6-10] light emission, even at sub-lifetime scales.[9-10]

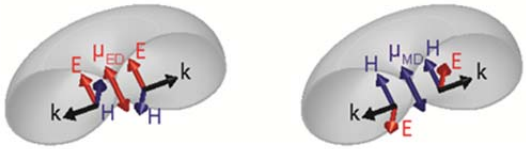

Figure 1: Schematics of electric and magnetic dipole emission in a homogenous environment.

ENERGY-MOMENTUM SPECTROSCOPY

When light is emitted by an electronic system, it radiates into specific optical modes within the local environment that are defined by energy, momentum, and polarization. Thus, in addition to its energy spectrum, light emission also has a distinct momentum spectrum. Light is not generally emitted out equally into all directions, but rather exhibits a characteristic radiation pattern. This radiation pattern encodes fundamental information about the specific quantum transitions from which it originates. Projecting this 2D radiation pattern (obtained by imaging the back-focal-plane of a microscope objective) onto the entrance slit of an imaging spectrograph yields an empirical dispersion diagram (i.e. an "energy-momentum spectrum") that shows how light is characteristically emitted into different angles, wavelengths, and polarizations.[2]

By comparing this energy-momentum spectrum with analytical theory, we can identify the strength, order, and orientation of different optical transitions.[2-6] For example, we initially developed energy-momentum spectroscopy to directly quantify the multipolar contributions to light emission from electric dipole (ED) and magnetic dipole (MD) transitions in lanthanide ions.[2,5,6] Based on interference effects near dielectric interfaces, ED and MD transitions may be readily identified from their distinct angular emission patterns (as shown in Figure 2e). Through detailed comparison of the energy-momentum spectra (Figure 2a & 2b) with theory, we were able to decompose light emission into its ED and MD contributions (Figure 2c) and measure the intrinsic ED and MD spontaneous emission rates (Figure 2d).

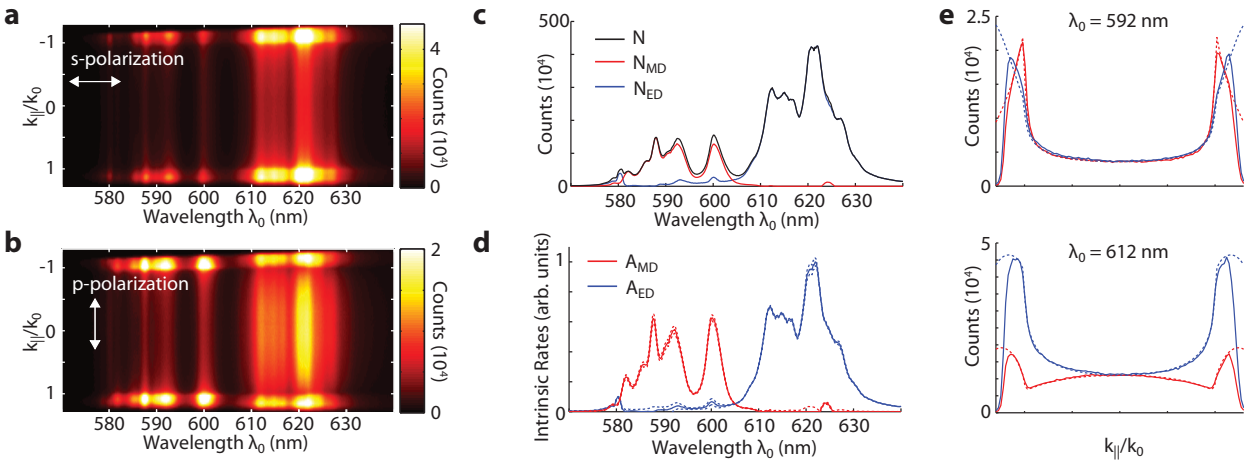

Figure 2. Quantifying the ED and MD transitions in Eu^{3+}:Y_2O_3 by energy-momentum spectroscopy[2]. (a,b) Experimental energy-momentum spectra for s- and p-polarization, respectively. **(c)** Integrated energy spectrum showing the total observed emission (N, black line) decomposed into the contributions from ED (N_{ED}, blue line) and MD (N_{MD}, red line) transitions. **(d)** Spectrally-resolved intrinsic emission rates, A_{ED} (solid blue line) and A_{MD} (solid red line), deduced from analysis of the energy-momentum spectra together with their 95% confidence intervals (dashed lines). **(e)** Momentum cross-sections for two representative wavelengths showing strong agreement between experimental data (solid lines) and theoretical fits (dashed lines) for both s-polarization (blue) and p-polarization (red). The measured momentum distribution for emission at 592 nm shows that this transition is predominantly magnetic (MD = 90.8 ± 2.0%), whereas 612-nm emission is almost purely electric (ED = 99.0 ± 1.0%).

Using energy-momentum spectroscopy, we have established light emission from the magnetic dipole transitions in lanthanide ions as a novel way to access the magnetic component of the optical field.[2] By decomposing luminescence into parts originating from ED and MD transitions, we have been able to measure the intrinsic spontaneous emission rates (Einstein A coefficients) and to use quantum transitions to independently probe vacuum fluctuations in the electric and magnetic field. We have extended our experimental method to directly measure the electric and the magnetic local density of optical states, as shown in Figure 3. These results reveal a new tool for nano-optics: an atomic-size quantum emitter that interacts with the magnetic component of light and therefore allows the study and exploitation of enhanced magnetic light-matter interactions at the nanoscale.

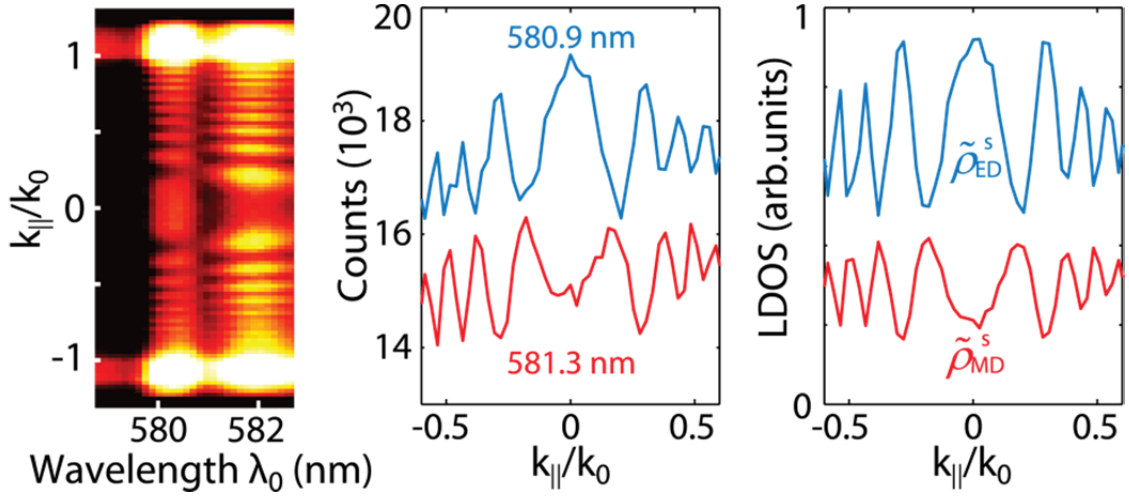

Figure 3: Measuring the electric and magnetic local density of optical states with ED and MD transitions[2]. (Left) Close-up of the back-focal-plane spectra from Eu^{3+} doped Y_2O_3 thin film near a planar interface highlighting the differences between the emission of the ED transition centered at 580.8nm and the MD centered at 582.2nm. (Center) Momentum cross sections near 581.1nm visualizing the different symmetry of ED and MD emission. (Right) Electric and magnetic local density of optical states (LDOS) near 581.1nm deduced from the momentum cross sections.

SUB-LIFETIME MODULATION OF EU^{3+}:Y_2O_3 EMISSION

We have demonstrated how the selective enhancement of electric and magnetic dipole transitions can be used to tune emission spectra, [7,8] even at sub-lifetime scales.[9] Figure 4 shows a pulsed excitation experiment wherein a scanning mirror was used to modulate emission between spectrally distinct electric dipole (ED) and magnetic dipole (MD) transitions in Eu^{3+}. Based on the mirror position, we can selectively enhance either ED or MD emission. The clear oscillations on the decay tail show that this modulation occurs at time scales faster than the lifetime limit.

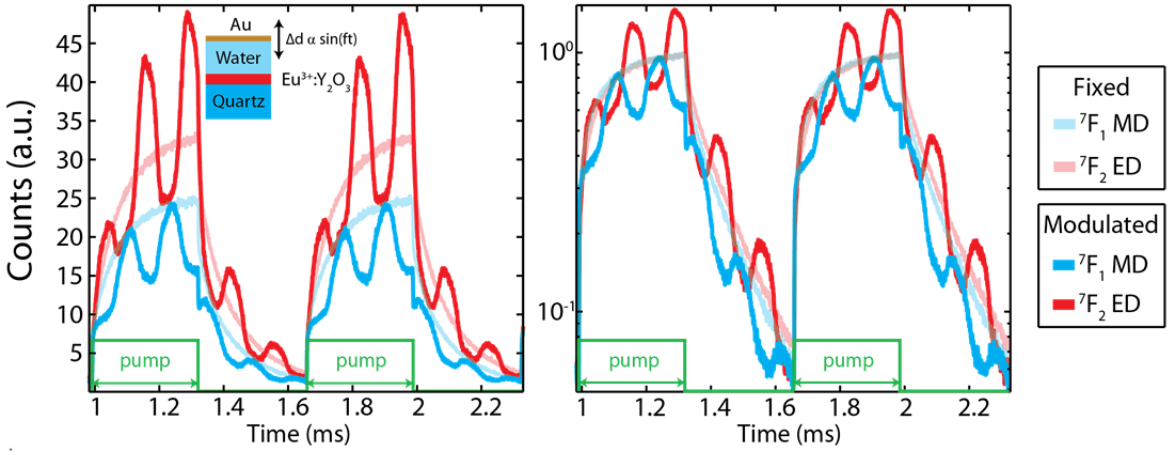

Figure 3: Time-Resolved Photoluminescence Confirming Dynamic Spectral Modulation at Sub-Lifetime Scale[9]. (Left) Linear and (right) log scale plots of the ED and MD emission for fixed and moving mirror cases. When the mirror position is fixed, the $^5D_0$ excited state of Eu^{3+} decreases as a single exponential with a lifetime of 108μs (shown in light red and blue lines). However, when the gold mirror position is dithered using piezoelectric crystal at a frequency of 7.5 kHz, the local density of optical states for the ED and MD transitions are dynamically modulated. This allows for the selective enhancement of either ED or MD emission within the excited state lifetime (shown in the dark red and blue lines) by the multiple oscillations after the end of each pump pulse.

FREE ION CALCULATIONS

We have performed detailed quantum-mechanical calculations in the intermediate coupling regime to identify strong magnetic dipole emission lines for future experimental studies.[1] While many magnetic dipole absorption lines have been previously identified, only a few magnetic dipole emission lines are widely known. Therefore, we calculated all the magnetic dipole transitions for the complete trivalent lanthanide series using a detailed free-ion Hamiltonian including, electrostatic and spin-orbit terms as well as two-body, three-body, spin-spin, spin-other-orbit, and electrostatically correlated spin-orbit interactions. These calculations predict a large number of strong magnetic dipole emission lines throughout the visible and near-infrared spectrum (as shown in Figure 5), including many at longer operating wavelengths that are ideal for experimental integration with plasmonics antennas and waveguides.

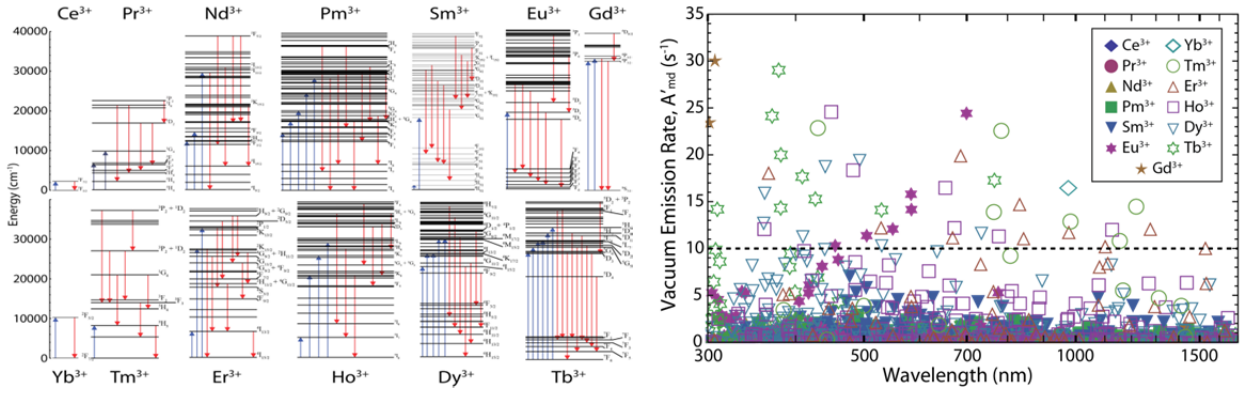

Figure 5. Magnetic dipole transitions in trivalent lanthanide ions[1]. (Left) Energy level diagrams highlighting strong magnetic dipole mediated absorption (blue) and emission (red) lines for all ions in the trivalent lanthanide series. (Right) Calculated emission rates for all MD emission lines.

MAGNETIC DIPOLE EMISSION OF $Dy^{3+}:Y_2O_3$ AND $Tm^{3+}:Y_2O_3$

We characterized the emission rates for multiple transitions in Dy^{3+} and Tm^{3+} doped Y_2O_3.[5] These lines could play important roles in various applications, including imaging and energy based upconversion processes. While these emission lines were previously calculated to have large spontaneous emission rates,[1] we find that the MD contribution to the overall emission is low. Further, we report that the overlapping $^4F_{9/2}\rightarrow{}^6F_{11/2}$ and $^4F_{9/2}\rightarrow{}^6H_{9/2}$ transitions in Dy^{3+}, centered at 750 nm, as well as the $^1G_4\rightarrow{}^3H_5$ and $^3H_4\rightarrow{}^3H_6$ transitions in Tm^{3+} show the greatest MD contributions, and thus provide the best pathways for future study. Other transitions in Dy^{3+} and Tm^{3+} were also characterized, but we find that their MD contributions in Y_2O_3 are relatively low. Though several of these emission lines have low MD contributions, they operate in the NIR where resonant structures are easier to fabricate and characterize. By integrating Dy^{3+} and Tm^{3+} with resonant nanostructures, one could enhance and engineer these MD emission lines to better leverage upconversion processes for use in energy harvesting and biological imaging.

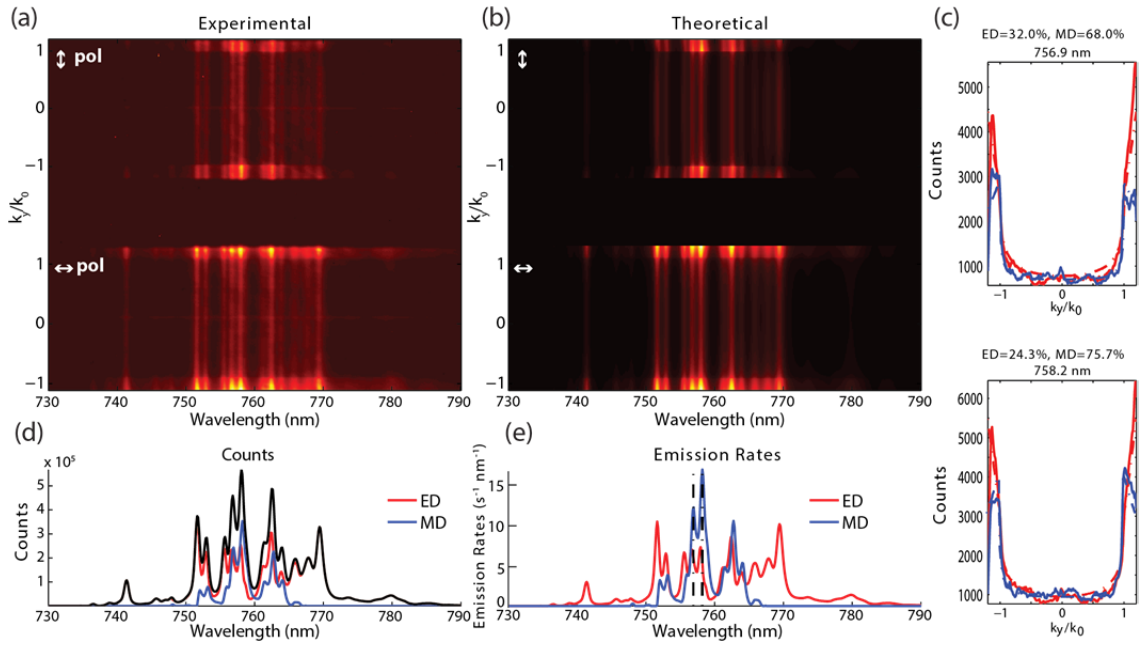

Figure 6: **Energy-momentum spectra of $^4F_{9/2}\rightarrow{}^6F_{11/2}$ and $^4F_{9/2}\rightarrow{}^6H_{9/2}$ transitions in $Dy^{3+}:Y_2O_3$ [5].** (a) Polarized experimental data and (b) corresponding fits. (c) Representative polarized cross-sections of experimental data (solid) and theoretical fits (dashed) at 756.9~nm, 32.0\% ED and 68.0\% MD, and 758.2~nm, 24.3\% ED and 75.7\% MD. Vertical polarization is shown in blue and horizontal polarization is shown in red. (d) Total (black) counts for ED (red) and MD (blue) emission. (e) Intrinsic emission rates for ED (red) and MD (blue) emission. The wavelengths of each cross-section are marked with dashed black lines. The white arrows in (a,b) denote polarization.

SPECTRAL TUNING OF ERBIUM LIGHT EMISSION BY SELECTIVE ED/MD ENHANCEMENT

We have demonstrated significant spectral tuning of erbium light emission near the 1.5 μm telecommunication band by selective enhancement of the electric and magnetic dipole contributions to the Er^{3+} $^4I_{13/2}\rightarrow{}^4I_{15/2}$ transition.[6] This work began by experimentally quantifying

the contribution of magnetic dipole (MD) transitions to the near-infrared light emission from trivalent erbium-doped yttrium oxide (Er^{3+}:Y_2O_3). Using our energy-momentum spectroscopy technique, we demonstrated that the $^4I_{13/2} \rightarrow {}^4I_{15/2}$ emission has a strongly mixed nature, with nearly equal contributions from electric dipole (ED) and MD transitions that exhibit distinct emission spectra. We then showed how these distinct spectra, together with the differing local density of optical states (LDOS) for ED and MD transitions, can be leveraged to tune Er^{3+} emission in nanostructured environments.

Note that, within any inhomogeneous or structured environment, the electric LDOS and the magnetic LDOS always differ and generally exhibit an inverse relationship. This difference in the LDOS is what allowed us to quantify ED and MD transitions by their radiation patterns. More importantly for active devices, the different LDOS experienced by ED and MD emitters can also be exploited to selectively direct emission into different transitions. Figure 7 illustrates how simple modifications to the local optical environment can be used to tune the emission of Er^{3+}:Y_2O_3 by altering the ratio of ED to MD emission in far-field spectra. Figure 7a shows normalized experimental emission spectra for the same Er^{3+}:Y_2O_3 emitter layer as a function of distance, *d*, from a gold mirror. (As shown in the inset of Figure 7d, these measurements were obtained by fabricating a staircase-like structure comprised of twenty-two different thickness spacer layers of Y_2O_3, in ~33 nm intervals from 0 to 697 nm, by masked electron-beam evaporation.) The observed experimental results in Figure 7a are in close agreement with our theoretical predictions for the modified emission spectra shown in Figure 7b. This agreement is highlighted by the comparison of the experimental and theoretical spectra at two representative

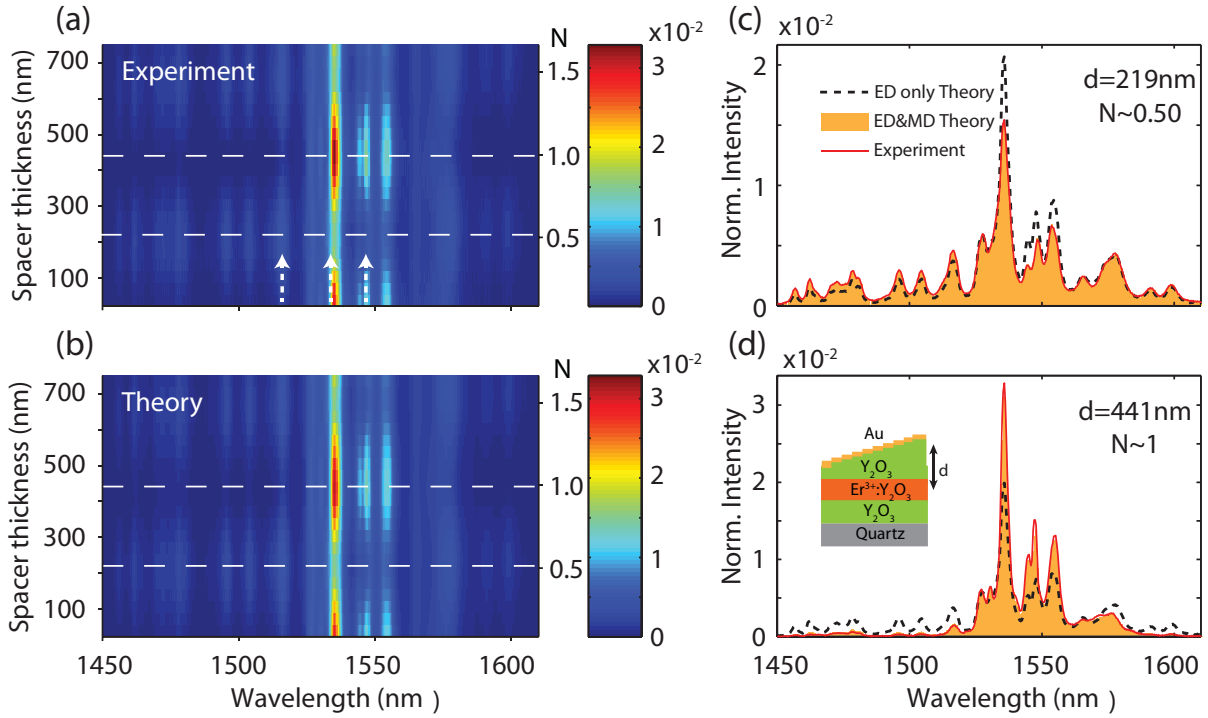

Figure 7. Modified emission spectra of Er^{3+}:Y_2O_3 near a gold mirror[6]. (a) Normalized experimental spectra versus spacer layer thickness; (b) Normalized theoretical spectra predicted by the three-layer structure self-interference model combining the intrinsic ED and MD emission rates. (c) and (d) are the extracted spectra from (a) and (b) along the white dashed lines at d = 219 and 441 nm. The theoretical normalized spectra (shaded orange region) accurately match the experimental normalized spectra plotted in red lines. For comparison, the predicted spectra by ED only model (black dashed lines) are also drawn here.

thicknesses in Figure 7c and 7d. These results clearly confirm that the observed spectral modifications can be accurately predicted by combining the measured ED and MD emission rates with a simple three-layer model for the LDOS. Importantly, these results also demonstrate that the observed spectra near the 1.5 μm telecommunication band can be tuned to resemble almost pure emission from either ED or MD transitions.

ALL-OPTICAL MODULATION OF ER^{3+} LIGHT EMISSION

Building on these results, we have realized a modulated 1550 nm source that leverages the refractive index modifications associated with the vanadium dioxide (VO_2) phase-change to dynamically control ED and MD emission from the near-infrared Er^{3+} $^4I_{13/2} \rightarrow {}^4I_{15/2}$ telecom line.[10] VO_2 is a strongly correlated material that undergoes a first order insulator-to-metal transition (IMT) when subjected to an appropriate external stimulus. Such an IMT can be induced thermally (around 68°C), electrically, optically, magnetically, or mechanically, and it has been reported to be as fast as hundreds of femtoseconds (when induced by femtosecond laser pulses). This transition has attracted significant interest from the photonics community, because this phase change is associated with a rapid modification in the optical properties of VO_2. As a consequence, both the refractive index and extinction coefficient are significantly modified.

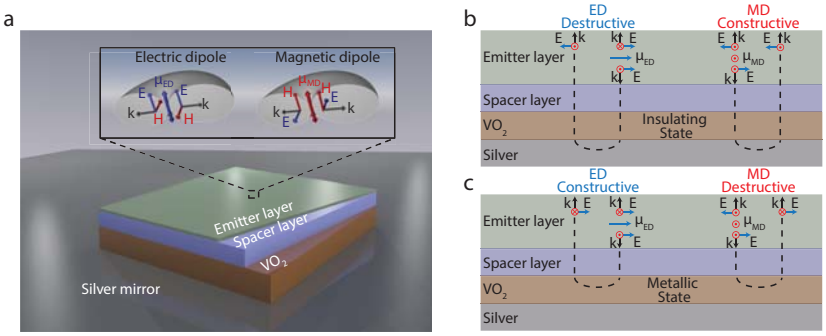

Figure 8. Sketch of the sample and illustration of LDOS modulation[10]. (a) Sketch of the sample. Inset shows the polarization symmetries of electric and magnetic dipole radiation. (b) and (c) illustrate the interference of ED and MD emission processes when the VO_2 layer is in the insulating and metallic phases, respectively.

By optically switching a VO_2 layer, we demonstrated modulation of Er^{3+}:Y_2O_3 emitters at sub-lifetime scales. To this end, the multilayer structure shown in the inset of Fig. 8a was designed such that the emitter layer has a high magnetic LDOS when VO_2 is in the insulating state, but switches to a high electric LDOS when VO_2 is in the metallic state, as shown in Fig. 8b,c. We demonstrated this experimentally by continuously pumping erbium with a 532 nm laser and using a 1064 nm control beam to induce the IMT without affecting the population of the Er^{3+} excited state. As shown in Fig. 9, the Er^{3+} emission spectrum switches from predominantly MD (red) to predominantly ED (blue) when the control beam is on. Additionally, we have demonstrated that such all-optical direct modulation can be leveraged to switch light emission faster than the lifetime of the emitter, as shown in the right panel of Fig. 9. Note these results were limited by the speed of the acousto-optic modulator used to generate the control pulses. Theoretically though, dynamic LDOS modulation is only limited by retardation effects, and therefore, the modulation speed of this technique could approach the ultrafast timescales of the IMT.

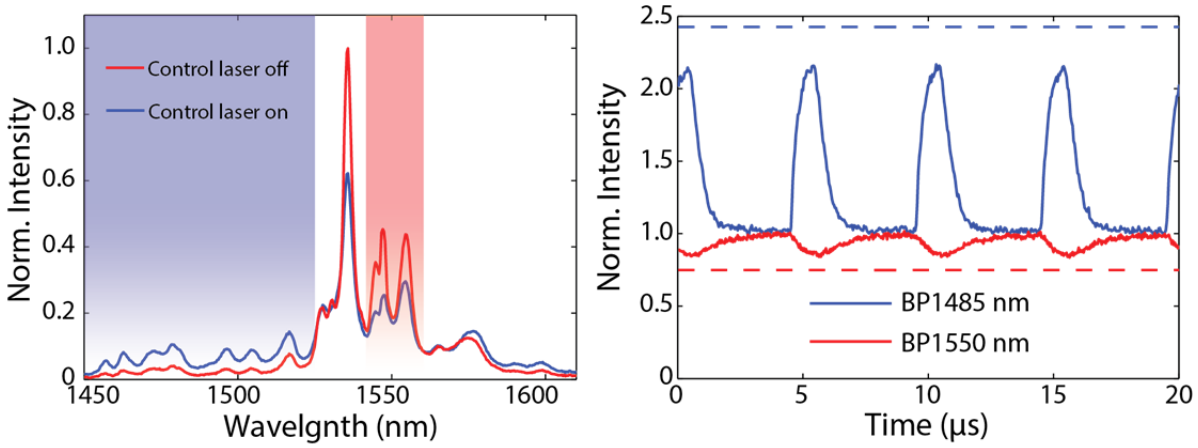

Figure 9. Sub-lifetime modulation of Er^{3+}:Y_2O_3[10]. (**left**) ED (blue) and MD (red) contributions to the emission of Er^{3+}:Y_2O_3 obtained from energy-momentum spectroscopy data on quartz substrate. (**right**) Emission spectrum of Er^{3+}:Y_2O_3 in layered device structure that switches from predominantly MD (red) to predominantly ED (blue) when 1064nm control beam induces the insulator-to-metal transition in VO_2.

WIDE-ANGLE ENERGY-MOMENTUM SPECTROSCOPY

More recently, we extended our energy-momentum spectroscopy technique to a broader range of emitters by removing the entrance slit on the imaging spectrograph and adding a Wollaston prism. Together, these modifications allow for the simultaneous acquisition of the full momentum and spectral distribution of light emission in a single measurement without the need for bandpass filters or scanning optics (Figure 10a). By leveraging the sparse nature of the multipolar basis, we were able to develop computational techniques that allowed us to extract meaningful information from these convolved data sets (e.g., the multipolar emission rates, see Figure 10e,f). Since this technique images the full radiation pattern at every wavelength in the measurement domain (e.g. Figure 10d), we're no longer restricted to isotropic emitters but instead can determine the wavelength dependent momentum distribution for emitters of arbitrary orientation and multipolar order (e.g., near optical antennas). Moreover, this technique also ensures that the etendue of the optical system is only limited by the collection efficiency of the microscope objective, thus allowing for the characterization of emitters that would otherwise be too dim (e.g., single quantum emitters such as nitrogen-vacancy and silicon-vacancy centers in diamond).

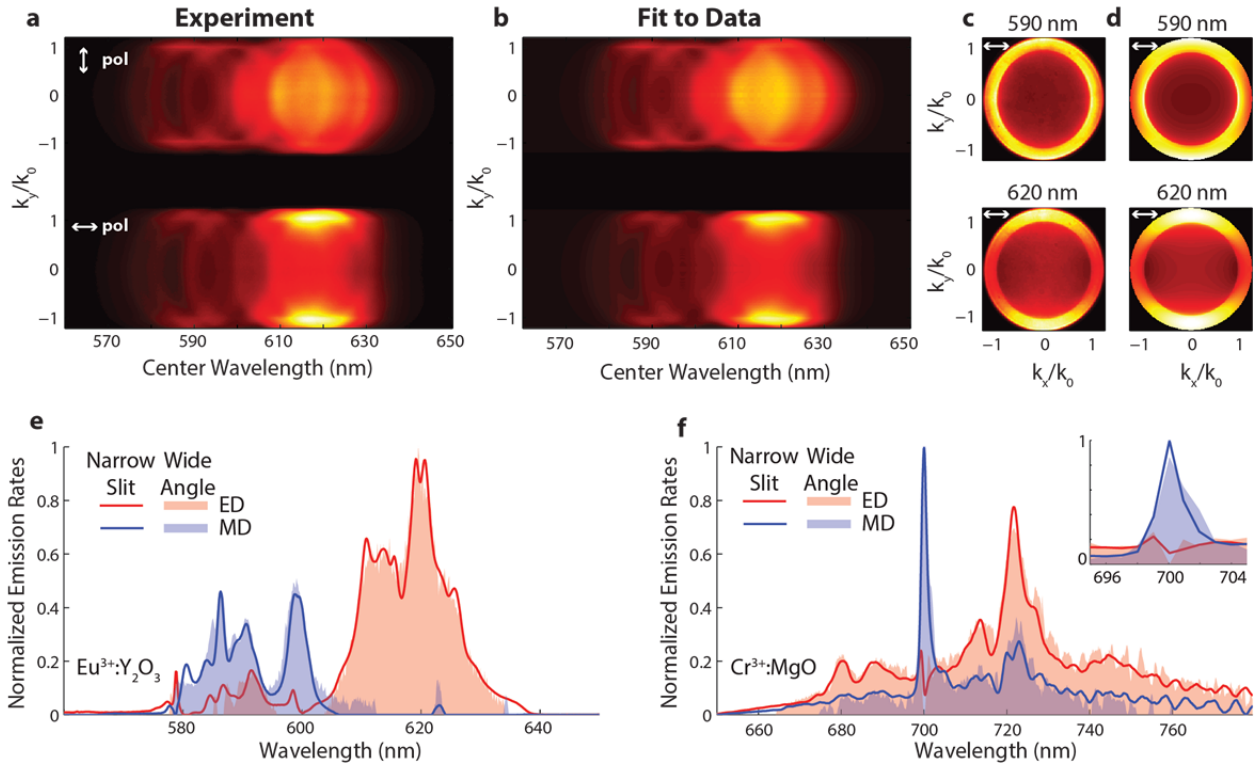

Figure 10.Wide-angle energy-momentum spectroscopy of Eu^{3+}:Y^2O^3 and Cr^{3+}:MgO[3] . (a) Experimental wide-angle data and (b) corresponding theoretical fits. (c) Experimental polarized 2D radiation patterns using 590±10 and 620±15 nm bandpass filters and (d) corresponding 2D radiation patterns extracted from wide-angle fits. White arrows denote polarization. Comparison of narrow-slit and wide-angle energy-momentum spectroscopy for (e) Eu^{3+}:Y^2O^3 and (f) Cr^{3+}:MgO. Extracted ED (red) and MD (blue) normalized emission rates from measurements both with (line) and without (shaded area) a ~10 μm entrance slit. The inset of (b) highlights the inverted symmetry of ED and MD emission at the ZPL near 700 nm.

EARLY CAREER AWARD IMPACT AND RELATED ACTIVITY, AWARDS, AND HONORS

In addition to the specific research accomplishments described above, this Presidential Early Career Award for Scientists and Engineers (PECASE) was instrumental in helping establish the PI's independent research group at Brown. This early career award helped to directly and indirectly support the work of 1 postdoctoral scholar and 5 graduate students (including 3 Ph.D. students who have successfully defended their doctorates and 2 others who are likely to defend within the following year). The discoveries made through this PECASE program have also helped serve as springboards for other successful research programs including an ongoing AFOSR Multi-University Research Initiative (MURI) on quantum metamaterials and a new NSF program on direct electrical modulation of light emission at sub-lifetime speeds as well as a completed AFOSR project on a cavity-free, matrix-addressable quantum dot architecture for chip-scale photonics. Based on the research results presented here, the PI has received a number of prestigious awards and honors. In 2010, the PI was named the Manning Assistant Professor of Engineering, an endowed junior faculty chair at Brown, and became a Fellow of the National Forum on the Future of Liberal Education. In 2011, he won the School of Engineering Dedicated Faculty Award. In 2012, he received the Henry Merritt Wriston Fellowship, gave a Dow Foundation Distinguished Lecture at the University of California, Santa Barbara, and became lead PI of the AFOSR-sponsored Quantum Metaphotonics & Metamaterials MURI program. In

2014, he received a lifetime award as an Outstanding Referee of the American Physical Society and was an invited plenary speaker at the triennial International Conference on Luminescence & Optical Spectroscopy of Condensed Matter.

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